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CARBIDE AND CARBON CHEMICALS CORPORATION

ENGINEERING DEVELOPMENT DIVISION

THEORETICAL ANALYSIS DEPARTMENT

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Report No.: A-4146
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Date: March 20, 1947

CALCULATION OF THE U^{234} GRADIENT IN THE GASEOUS DIFFUSION PLANT

Written by G. A. Garrett

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CARBIDE AND CARBON CHEMICALS CORPORATION

ENGINEERING DEVELOPMENT DIVISION

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To: S. Cromer

Report No.: A-4146
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Date: March 20, 1947

CALCULATION OF THE U^{234} GRADIENT IN THE GASEOUS DIFFUSION PLANT

Written by: G. A. Garrett

Abstract

An approximate method is described for calculating the steady-state U^{234} concentration at any point in the plant. The method consists of making two calculations, as follows:

(1) Making the usual productivity calculation and regarding all concentrations as concentrations of U^{234} plus U^{235}.

(2) Making a second calculation for determining the fraction of each concentration calculated in (1) which is U^{234}.

The steady-state U^{234} concentration of the product for current operations as calculated by this method is 2221501%. Laboratory analyses on March 4th gave the product concentration of U^{234} as 2223001%, and similar analyses over a period of time show that the U^{234} gradient probably had not attained steady-state on this date, since the U^{234} concentration of the product was decreasing gradually with time.

Curves are presented showing the calculated U^{234} gradient in the plant and that obtained from laboratory analyses on March 4th. The latter gradient is consistently higher than the former by about 11% of the values.

The calculations are based upon an assumed value of 1:18000 for the concentration of U^{234} in normal feed material.

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Introduction

In all equations and calculations of the Theoretical Analysis Department heretofore the process gas has been treated as a mixture of two components, $U^{235}F_6$ and $U^{238}F_6$, whose molecular weights are 349 and 352 respectively. The presence of the component $U^{234}F_6$ of molecular weight 348 has been neglected since, although the $U^{234}F_6$ is enriched in the cascade by a greater factor than the $U^{235}F_6$, the concentration of the U^{234} is still so small that it is usually negligible insofar as the gaseous diffusion plant is concerned. However, during operations at high product concentrations, it is desirable to be able to determine the 234 content of the product and at various points in the cascade.

The similarity between the mathematical treatment of a section of the gaseous diffusion plant and that of a fractionisation column has been described in previous reports from this department. Precise mathematical treatment of fractionization columns operating on materials other than a binary mixture is extremely tedious, and usually involves an iterative tray-to-tray type of computation. Similar methods are practically hopeless when applied to the diffusion cascade where the number of stages runs into thousands. Fortunately, however, the fact that the U-234 concentration is always small compared to the U-235 concentration makes it possible to develop an approximate method for a rapid calculation of U-234 concentrations throughout the cascade. It is the purpose of the present report to present this approximate method for calculating U-234 concentrations.

Conclusions

Figure 2 presents the steady-state U-234 gradient for the K-25 Plant as calculated by the method described herein, and the U-234 gradient obtained by laboratory analyses of samples taken March 4, 1947. The laboratory analyses of the product over a period of time show that the steady-state gradient probably has not yet been reached, since the U-234 concentration of the product is still decreasing very gradually. The steady-state concentration of U-234 in the product is calculated to be 2221501%. The laboratory analysis on the above date was 2223001%. A value of 1:18000 for the concentration of U-234 in normal feed material was used in the calculations. The value for the natural abundance of U^{234} apparently is not accurately known. The computed values in Figure 2 are approximately proportional to the assumed value for the natural abundance.

Derivation of Calculating Procedure

The formulas used in productivity calculations for a section in the enricher are obtained from solution of the differential equation

$$\frac{dx}{dn} = \psi x(1-x) - \frac{P}{L}(y_p - x) \qquad (1)$$

where
- x is the light component concentration of the tails stream at stage n (mol %)
- y_p is the light component concentration of the product (mol %)
- L is the upflow rate (lb. mols/day)
- P is the product rate (lb. mols/day)

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If we use subscripts as illustrated in Figure 1,

$$\psi = \frac{y_i - x_i}{x_i (1 - x_i)} \tag{2}$$

A general theoretical expression, applicable to all barriers, relating psi and operating pressures is not known. However, it is generally believed that for separation of two isotopes, the relationship between ψ and pressure has the form

$$\psi = \left[\sqrt{\frac{M_2}{M_1}} - 1\right] g(P_0, P_1) \tag{3}$$

where $g(P_0, P_1)$ is some function of the high-and low-side pressures, and M_2/M_1 is the ratio of molecular weights of the isotopes.

Now at any point in the cascade the average molecular weight of the $U^{234}F_6$ and $U^{235}F_6$ is not sensibly different from that of the $U^{235}F_6$ itself. This statement suggests the following approach to the three-isotope problem:

(1) Consider the symbols x and y_p in Equation 1 and the x's and y's of equation 2 as applying to concentrations of the mixture of $U^{235}F_6$ and $U^{234}F_6$ at the appropriate points. In other words, in this phase of the problem we consider the $U^{234}F_6$ and $U^{235}F_6$ together as a single component.

(2) Develop additional equations for determing the fractions of x and y_p which are $U^{234}F_6$.

We now shall develop these additional equations for use in this second phase of the problem. Referring to Figure 1, and using the nomenclature given therein, a material balance on the $U^{234}F_6$ across the two streams joining stage i and stage i + 1 gives us the equation

$$L y_i w_i = (L - P) x_{i+1} z_{i+1} + P y_p z_p \tag{4}$$

We denote by the symbol φ the analogue to ψ, as applied to the enrichment of $U^{234}F_6$ in a mixture of $U^{234}F_6$ and $U^{235}F_6$. Thus

$$\varphi = \frac{w_i - z_i}{z_i (1 - z_i)} \tag{5}$$

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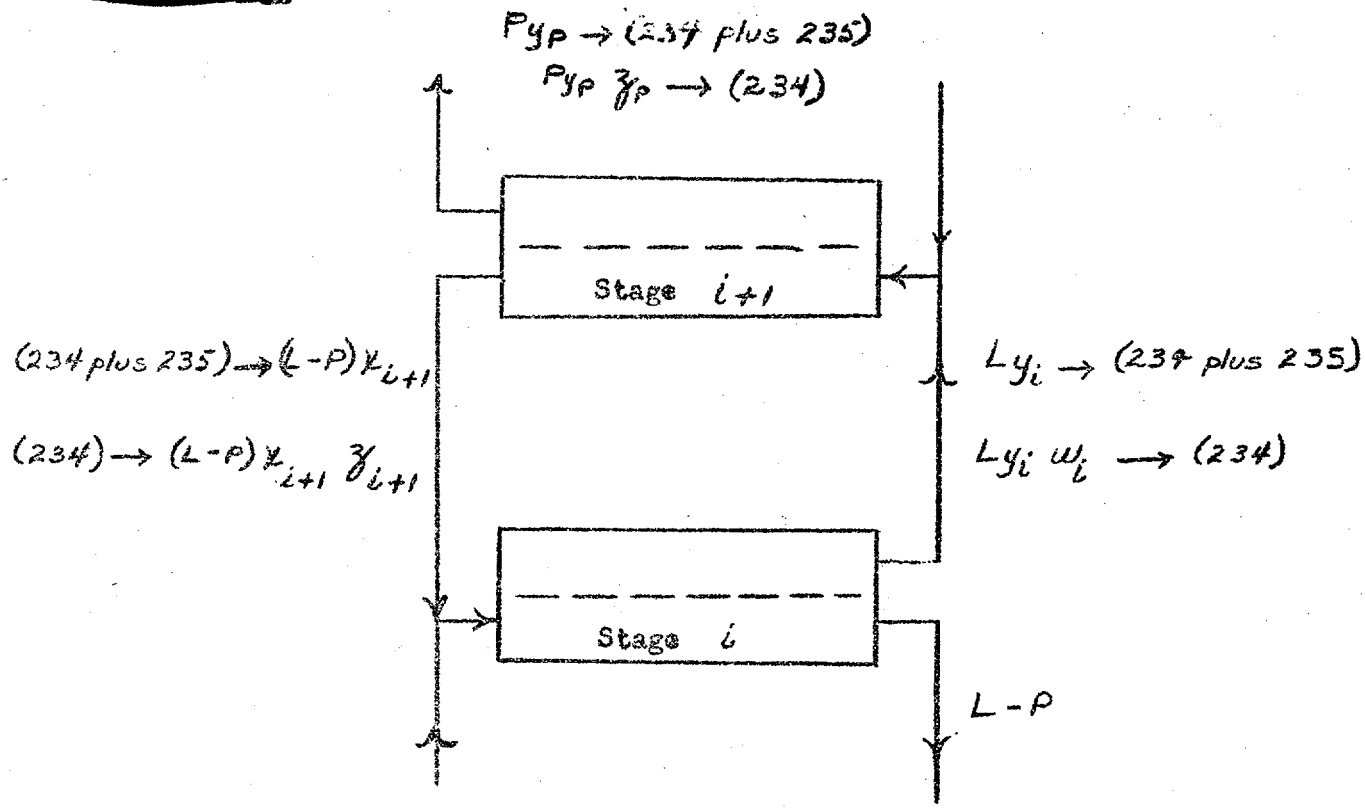
Product.



x_i = mol fraction of 234 plus 235 in the Uranium in the tails stream from stage i

z_i = mol fraction of x_i which is 234

y_i = mol fraction of 234 plus 235 in the Uranium in the heads stream from stage i.

w_i = mol fraction of y_i which is 234

y_p = mol fraction of 234 plus 235 in product

z_p = mol fraction of y_p which is 234

L = Flow rate in heads stream (lb. mols/day)

P = Product rate (lb. mols/day)

Figure 1.

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Assuming that Equation (3) holds and that φ obeys a similar relationship, we have

$$\varphi = \frac{\sqrt{\frac{349}{348}} - 1}{\sqrt{\frac{352}{349}} - 1} \cdot \psi = 0.3347\,\psi \tag{6}$$

By means of Equations (2) and (5), we obtain

$$y_i w_i = x_i z_i \left[1 + \varphi(1-z_i) + \psi(1-x_i) + \varphi\psi(1-x_i)(1-z_i)\right]$$

$$\cong x_i z_i \left[1 + \varphi(1-z_i) + \psi(1-x_i)\right] \tag{7}$$

Substituting (7) in (4), we have

$$x_{i+1} z_{i+1} - x_i z_i = \frac{1}{1-\frac{P}{L}} \left\{ x_i z_i \left[\varphi(1-z_i) + \psi(1-x_i)\right] - \frac{P}{L}(y_p z_p - x_i z_i) \right\} \tag{8}$$

Neglecting P/L, which is small compared to unity, in the denominator on the right and dropping subscripts as is conventionally done in deriving Equation (1), Equation (8) takes the form

$$\frac{d}{dn}(xz) = xz\left[\varphi(1-z) + \psi(1-x)\right] - \frac{P}{L}(y_p z_p - xz) \tag{9}$$

or

$$x\frac{dz}{dn} + z\frac{dx}{dn} = xz\left[\varphi(1-z) + \psi(1-x)\right] - \frac{P}{L}(y_p z_p - xz) \tag{10}$$

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Eliminating $\frac{dx}{dn}$ by means of (1), we obtain

$$\frac{dz}{dn} = \varphi z(1-z) - \frac{P_{y_p}}{Lx}(z_p - z) \qquad (11)$$

Equations (11) and (1) form a system of two differential equations which, if solved simultaneously, would lead to the determination of z and x at every point of interest. Efforts at finding useful formulas by a simultaneous solution were unfruitful. However, by replacing x in Equation (11) by $\bar{x}$, the average value of x for the section as obtained by the usual productivity calculation, it was found that the calculated results agreed very well with the plant gradient. This approximate formula, for a section in the enricher, is

$$\frac{dz}{dn} \cong \varphi z(1-z) - \frac{P_{y_p}}{L\bar{x}}(z_p - z) \qquad (12)$$

Analogous equations are obtained for stripping sections. By inspection of Equations (12) and (1) we note that Equation (1) is transformed into Equation "12) by the following changes

(1) Replacing x by z

(2) Replacing P by P_{y_p}

(3) Replacing L by $L\bar{x}$

(4) Replacing ψ by φ

Thus, it is seen that once an "x" productivity calculation has been made, then the "z" calculation can easily be made by making a second calculation after changes (2), (3) and (4) are made in all calculation sheets.

It should be borne in mind that the calculated x's are concentrations of $U^{234}F_6$ plus $U^{235}F_6$, and that the z's are fractions of the x's which are $U^{234}F_6$.

Figure 2 shows the calculated steady-state U^{234} gradient as computed by the method described above, and also the U^{234} gradient as obtained by laboratory analyses of samples taken on March 4, 1947. Also shown in Figure 2 is the U^{235} gradient obtained by subtracting the calculated U^{234} concentrations from the "x" gradient.

In view of the numerous approximations made in the derivation of this method, we shall review them at this point in order to point out the main uncertainties in the method. All approximations in the derivations are trivial with the exception of two, as follows:

(1) The use of $\bar{x}$ in place of x in Equation (12)

(2) The uncertainty in "φ".

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The effect of approximation (1) may be made as small as desired by breaking up the large sections into a number of sub-sections and treating each sub-section, using the $\bar{x}$ for the sub-section, in the same manner in which the sections are usually treated.

Figure 3 shows the "calculated" "x" gradient, the ratio of the concentration of $U^{234}F_6$ to that of the $U^{234}F_6$ and $U^{235}F_6$ mixture. This is approximately the same as the ratio of the U^{234} concentration to that of U^{235}. It is interesting to note that in some of the lower sections of the plant the ratio of U^{234} to U^{235} is slightly less at the top of the section than at the bottom. This means that, as far as the separation of U^{234} from U^{235} is concerned, these sections operate below the minimum reflux concentration. This also means that these sections enrich the isotope of medium molecular weight relatively higher than either of the other two. This fact suggests that it may be possible in the design of gaseous diffusion plants to provide for the enrichment or depletion of one isotope relatively more than the other isotopes.

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The formulas derived herein were set up for productivity calculations by W. G. Siedenburg. The "x" calculations were made by E. Usdin.

The U-234 Analyses of samples taken March 4th were analyzed by the Works Laboratory at the request of E. B. Carter. The data were supplied by Dr. F. W. Hurd.

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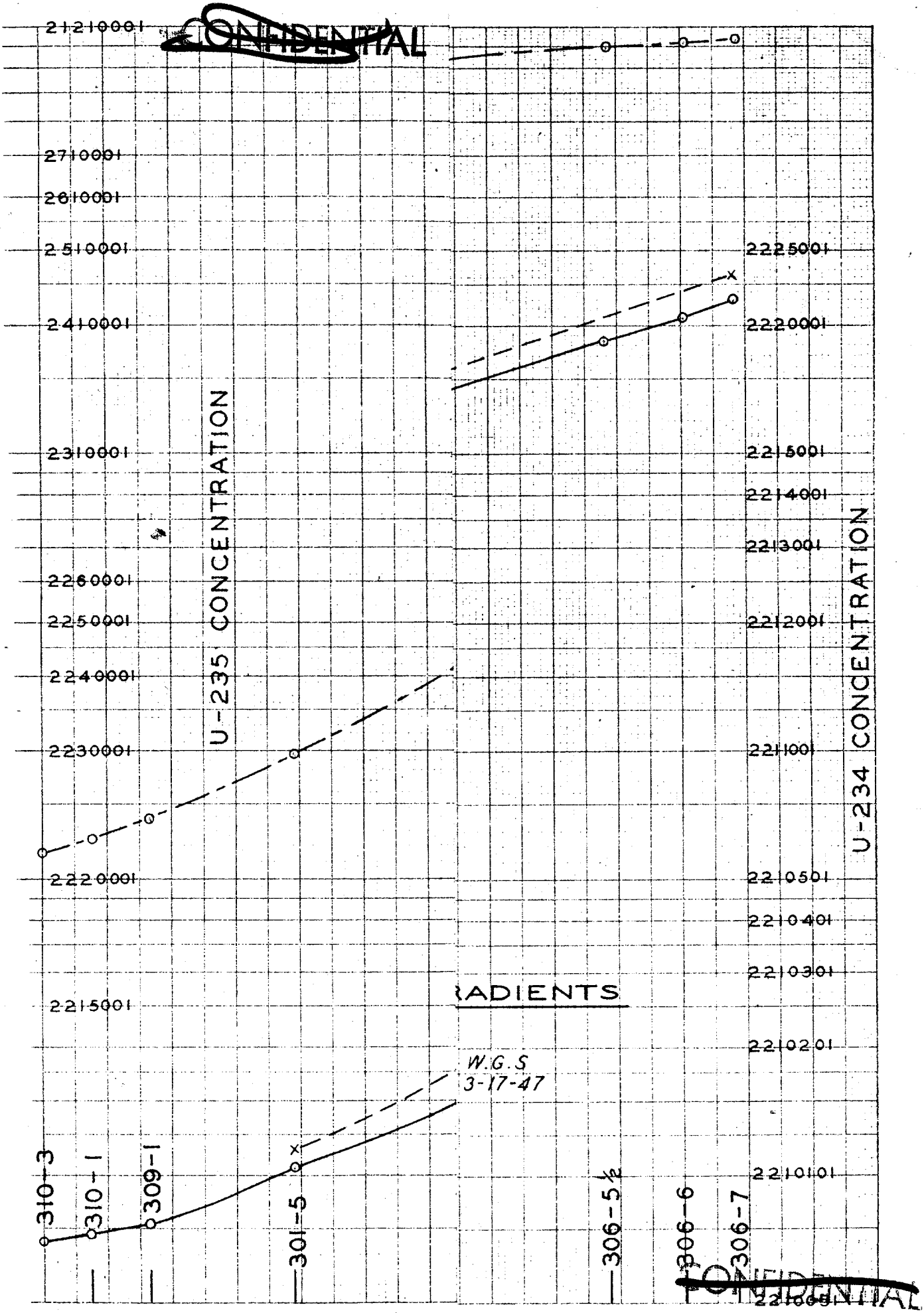
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FIGURE 2

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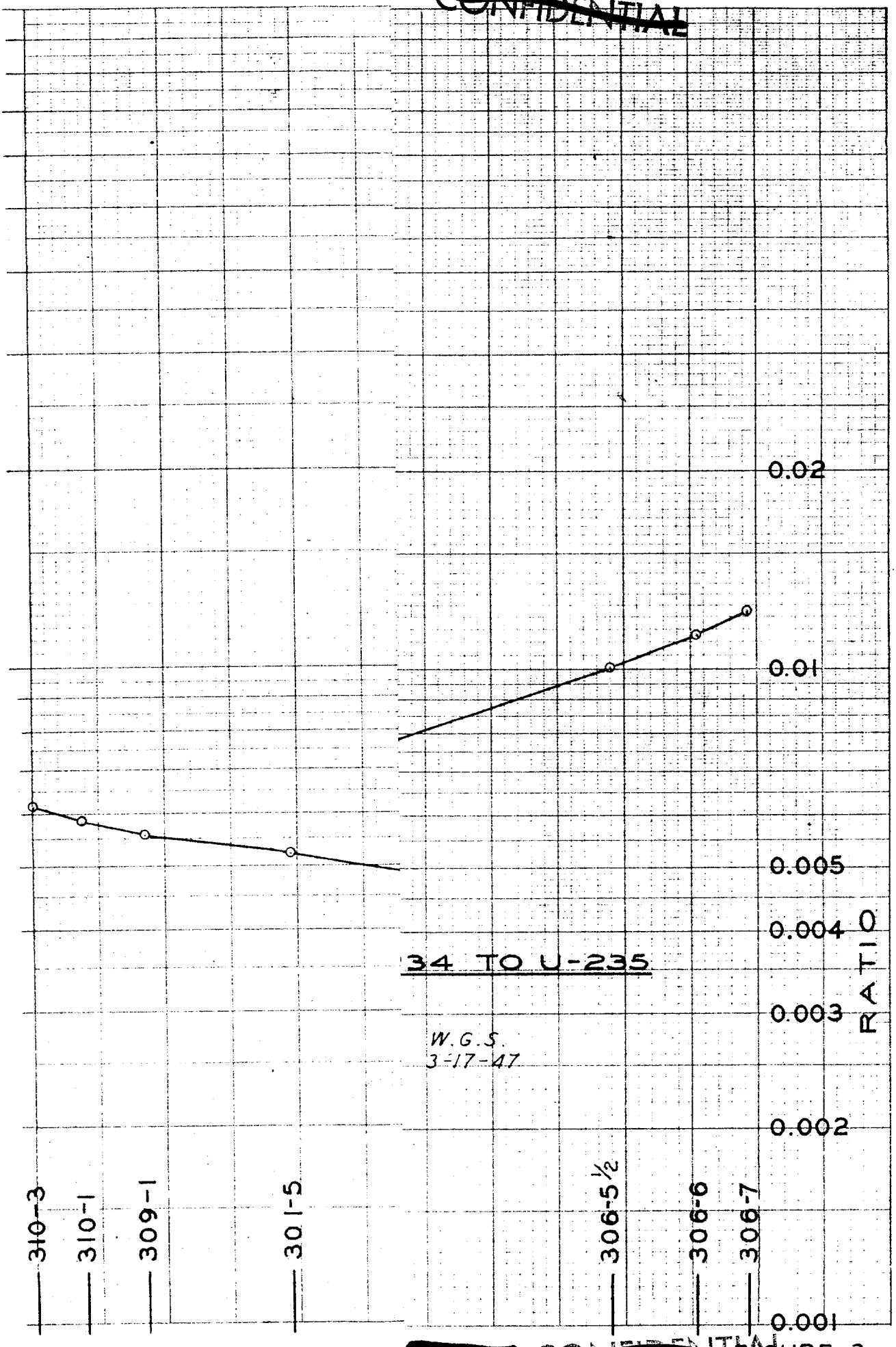

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FIGURE 3